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Title: Studies of Fission-Induced Surface Damage in Actinides Using Ultracold Neutrons

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Studies of Fission-Induced Surface Damage in Actinides Using Ultracold Neutrons

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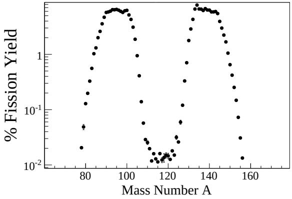
Fission-Induced Damage

Sputtering

- Fission event: 2 fragments, E∼100 MeV, A∼100
- Fast, heavy charged particles ⟶ ejection of atoms
- Damage to material surface

^{235}U fission fragments



Not well understood

- Underlying mechanism?
- Sputtered atoms per fission event?
- Damage to the material surface?
- Competing models?
- Quality of surface (oxide layer)?
- Sputtering from "deep" fissions (∼10 μm)?

Aging of Nuclear Materials

- Reactor fuel rods
- Satellites: thin film on batteries
- Stockpile stewardship

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Ultracold Neutrons

Class	Energy	Source
Fast	> 1 MeV	Fission reactions / Spallation
Slow	eV – keV	Moderation
Thermal	0.025 ev	Thermal equilibrium
Cold	μeV – meV	Cold moderation
Ultracold	≤ 300 neV	Downscattering

How cold is Ultracold?

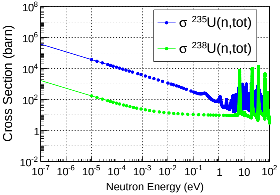
- Temperature < 4 mK
- Velocity < 8 m/s
- Usain Bolt ~ 12 m/s

UCN can be bottled

- Gravitational ($V = mgh$): 100 neV / meter
- Magnetic ($V = -\vec{\mu} \cdot \vec{B}$): 60 neV / Tesla
- Material $\left(V = \frac{2\pi\hbar^2 Nb}{m}\right) \begin{cases} {}^{58}\text{Ni}: & 335\ \text{neV} \\ \text{DLC}: & 250\ \text{neV} \\ \text{BeO}: & 250\ \text{neV} \\ \text{Cu}: & 170\ \text{neV} \end{cases}$

UCN-Induced Fission

Very high cross section: $\sigma \sim \frac{1}{v}$

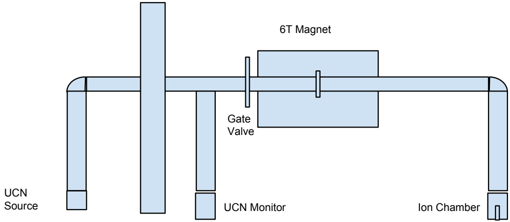


Cross Section (barns)			
UCN Energy	200 neV	300 neV	400 neV
^{235}U(n,tot)	2.64×10^{5}	2.16×10^{5}	1.87×10^{5}
^{238}U(n,tot)	1.17×10^{3}	9.57×10^{2}	8.29×10^{2}

Finely tune depth into material

UCN range in foil (μm)				
Comp.	% ^{235}U	200 neV	300 neV	400 neV)
DU	0.2%	118	144	191
NatU	0.7%	66	81	101
SEU	2%	31	38	45
LEU	5%	13	17	20
HEU	20%	4	4.5	5
	100%	0.8	0.9	1

UCN Source at LANSCE



UCN Source[1]

- UCN Source: 800 MeV proton beam + Tungsten target = CN
- CN downscatter in SD_2 crystal = UCN
- UCN Monitor = Normalize for fluctuations in UCN production

Detection

- Gate valve permits UCN entry to experiment
- 6 T magnet = near 100% polarization
- UCN drop through Al window into ion chamber

[1] Rev. Sci. Instrum. **84** 013304 (2013)

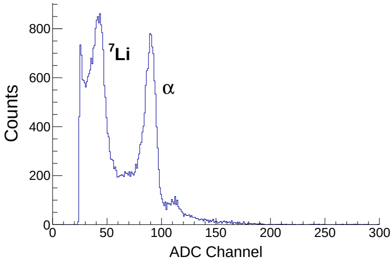
UCN Rate Normalization

UCN Beam Monitor[2]

- ^{3}He filled multi-wire proportional chamber
- $^3\mathrm{He} + \mathrm{n} \rightarrow \mathrm{p}$ (573 keV) + t (191 keV)
- 50% transmission through window into detector; 80% efficient

Baseline UCN Rates[3]

- Boron-coated cylindrical ion chamber, 1 barr argon
- $^{10}\mathrm{B} + \mathrm{n} \rightarrow \alpha + {}^7\mathrm{Li}$
- Near 100% efficient for UCN entering chamber
- Rate: 4.5kHz (for 125 Hz beam monitor rate)



[2]Nucl. Instrum. Meth. Phys. Res. A **599** 248 (2009)

[3]Nucl. Instrum. Meth. Phys. Res. A **691** 109 (2012)

Proof of Concept: Fission Rate

Experiment

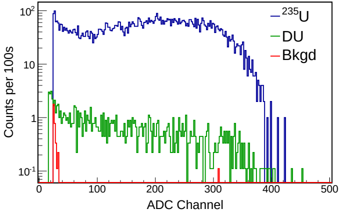
- Identical experimental setup
- Cylindrical ion chamber with boron coating removed
- Effect of UCN bottling?
- 200 mbarr argon: α's range out

^{235}U

- 2.2 cm diameter, 1 mm thick disk of HEU ($> 80\%$ ^{235}U)
- Rate: $(1.90 \pm 0.02) \times 10^{-2}$ fission/UCN

^{238}U

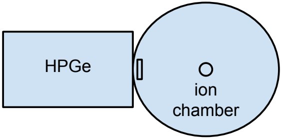
- 2.25 cm diameter, 1 mm thick disk of Depleted Uranium ($\sim 0.2\%$ ^{235}U)
- Rate: $(1.3 \pm 0.8) \times 10^{-4}$ fission/UCN



Neutron Capture Gammas

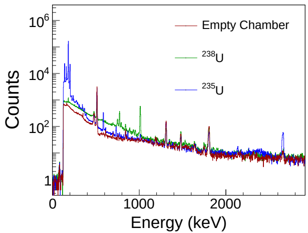
HPGe detector

- Calibration: ^{60}Co and ^{137}Cs gamma sources
- Goal: tag gamma, look for fission



Observed Spectra

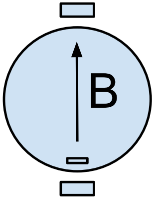
- Empty chamber with/without UCN: additional 480 keV line from residual Boron coating
- Decay gammas from ^{235}U/^{238}U observed; some additional lines
- No additional gamma lines with UCN?



Neutron Spin Dependence

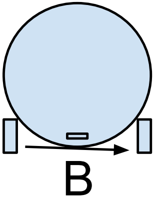
Neutron Polarization

- 6 T Magnet: near 100% UCN polarization
- Neutron spin aligned with field



Experiment

- Neodymium magnets installed on chamber: $\vec{B}$ field normal and parallel to surface
- $\sim$200G field normal to surface: $(1.92 \pm 0.02) \times 10^{-2}$ fission/UCN
- $\sim$50G field parallel to surface: $(1.94 \pm 0.02) \times 10^{-2}$ fission/UCN
- No magnets: $(1.90 \pm 0.02) \times 10^{-2}$ fission/UCN



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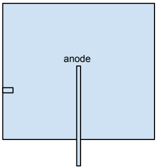
Sputtering

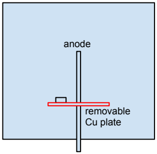
Evidence of UCN-induced sputtering?

- Installed ^{235}U for $\sim$20 minutes
- Exposed to UCN for $\sim$10 minutes
- Removed sample: small signal still observed!
- α rate = 2.63$\pm$0.07 Hz ($\sim 10^{17}$ atoms)

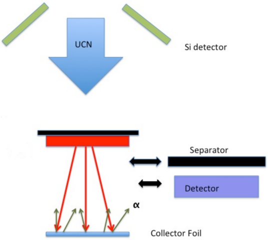


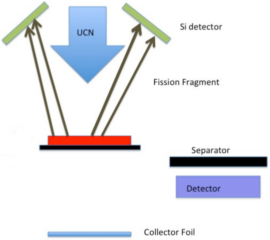
Check: No UCN exposure

- ^{235}U installed on removable copper plate: reduce chance of contamination
- Installed for $\sim$15 minutes, not exposed to UCN
- Removed copper plate with sample
- α rate = 0.78$\pm$0.04 Hz ($\sim 10^{16}$ atoms)
- Inconclusive: contamination? α-induced sputtering? chamber pumping/pressurizing?



Characterize Ejected Material





Important questions:

- How much comes off?
- Size distribution vs. depth/surface quality?
- Kinetics vs. depth?

Summary

First observation of UCN-induced fission

- Previously no fission data at these energies
- Determine relative cross-sections (e.g. vanadium sample)

Next: Confirm UCN-induced sputtering

- Sputtered rate as function of exposure time
- Better sample mounting: eliminate possibility of contamination
- Electropolish sample: clean, well-understood surface

Program Goals

- Characterize sputtered ejecta from various actinides
- Control fission depth via UCN energy: gravity/magnetic potentials
- Understand effect of depth and surface quality
- Examine different alloys, material layers